



# UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE  
United States Patent and Trademark Office  
Address: COMMISSIONER FOR PATENTS  
P.O. Box 1450  
Alexandria, Virginia 22313-1450  
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
09/555,544	08/01/2000	KWANG-LEONG CHOY	674505-2011	8846

20999 7590 04/13/2005

FROMMER LAWRENCE & HAUG  
745 FIFTH AVENUE- 10TH FL.  
NEW YORK, NY 10151

EXAMINER

VINCENT, SEAN E

ART UNIT	PAPER NUMBER
----------	--------------

1731

DATE MAILED: 04/13/2005

Please find below and/or attached an Office communication concerning this application or proceeding.

## Office Action Summary

Application No.

09/555,544

Applicant(s)

CHOY ET AL

Examiner

Sean E. Vincent

Art Unit

1731

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

### Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If the period for reply specified above is less than thirty (30) days, a reply within the statutory minimum of thirty (30) days will be considered timely.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

### Status

- 1) ☒ Responsive to communication(s) filed on 13 December 2004.
- 2a) ☐ This action is **FINAL**. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

### Disposition of Claims

- 4) ☒ Claim(s) 29-56 is/are pending in the application.
- 4a) Of the above claim(s) \_\_\_\_\_ is/are withdrawn from consideration.
- 5) ☐ Claim(s) \_\_\_\_\_ is/are allowed.
- 6) ☒ Claim(s) 29-56 is/are rejected.
- 7) ☐ Claim(s) \_\_\_\_\_ is/are objected to.
- 8) ☐ Claim(s) \_\_\_\_\_ are subject to restriction and/or election requirement.

### Application Papers

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on \_\_\_\_\_ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.  
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).  
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

### Priority under 35 U.S.C. § 119

- 12) ☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☒ All b) ☐ Some \* c) ☐ None of:
- ☐ Certified copies of the priority documents have been received.
  - ☐ Certified copies of the priority documents have been received in Application No. \_\_\_\_\_.
  - ☒ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

\* See the attached detailed Office action for a list of the certified copies not received.

### Attachment(s)

- 1) ☐ Notice of References Cited (PTO-892)
- 2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
- 3) ☐ Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08)  
Paper No(s)/Mail Date \_\_\_\_\_.
- 4) ☐ Interview Summary (PTO-413)  
Paper No(s)/Mail Date. \_\_\_\_\_.
- 5) ☐ Notice of Informal Patent Application (PTO-152)
- 6) ☐ Other: \_\_\_\_\_.

20

## DETAILED ACTION

### *Claim Rejections - 35 USC § 102*

1. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

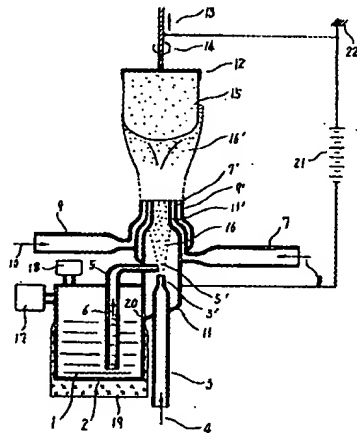
(b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

2. Claims 45-50 and 52-56 are rejected under 35 U.S.C. 102(b) as being clearly anticipated by Hitachi (JP 56-5337).

Hitachi's English abstract stated (with emphasis added):

The glass raw material 1 of a glass composition is atomized by means of a gas containing hydrogen from the nozzle 3', mixed with an oxidative gas from the pipe 9 and then burned, and **then the flame 16' is blasted against the target 12 to deposit the glass fine particles 15**. In this case, a DC voltage is applied between the negative voltage-nozzle 3' and the earthed target 12. Thus, the negatively charged glass fine particles with ionized gas coming out of the nozzle 3' collides with the earthed target 12 having a sign different from that of the glass fine particles rapidly and effectively to deposit the glass fine particles 15 of an uniform thickness in order to obtain the optical fiber mother material 12.

第 1 図



3. Hitachi's apparatus was identical in structure to the claimed apparatus as it included a first central outlet 11', a second outlet 9', a third outlet 7', a precursor supply 1, an electrical supply 21, an annular electrode 23, a positioner 13, and a burner (9', 11' and 7' form a burner face). Note MPEP 2114.

***Claim Rejections - 35 USC § 103***

4. The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office action.

5. This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).

6. Claim 51 is rejected under 35 U.S.C. 103(a) as being unpatentable over Hitachi in view of Japan 62-220376 (see abstract and figures).

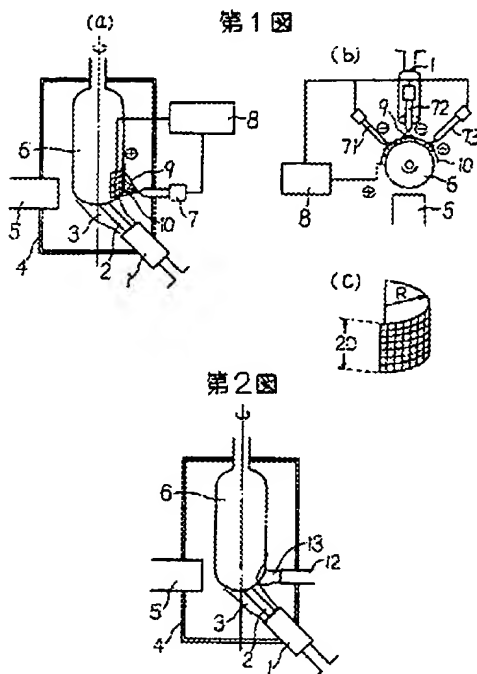
7. Hitachi did not teach a mesh disposed between the first outlet and the substrate. Japan '376 taught:

**PURPOSE:** To efficiently obtain the title preform without cracking by cooling a porous preform with an ion jet generated by a corona discharge, and simultaneously depositing fine glass particles on the preform at the time of producing the porous optical fiber preform by the axis formation in a vapor

Art Unit: 1731

phase.

**CONSTITUTION:** A corona-discharge electrode 7 is arranged above a fine glass particle synthesizing burner 1. A glass material and a combustion gas are supplied to the burner 1 to cause a flame hydrolysis reaction, the formed fine glass particles 3 are deposited on the surface of a starting material, and a porous optical fiber preform 6 is obtained. A negative voltage is simultaneously impressed on the corona-discharge electrode 7 and a positive voltage on a wire-mesh electrode 10 from a DC power source 8 to generate a corona discharge between both electrodes, and an ion jet 9 is produced. The porous preform 6 is cooled by the ion jet 9, the fine glass particles are simultaneously deposited, and the preform 6 is produced.



8. It would have been obvious to a person of ordinary skill in the art at the time the invention was made to use a wire-mesh electrode like that of Japan '376 in the arrangement of Hitachi because a mesh would have been equivalent to the electrode of Hitachi.
9. Claims 29-50 and 52-56 are rejected under 35 U.S.C. 103(a) as being unpatentable over Choy et al (WO 97/21848) in view of Blackwell et al (US 6312656).

Art Unit: 1731

10. Choy et al teaches apparatus and methods for depositing oxide or ceramic material on a substrate including delivering a stream of precursor droplets, applying an electric field between the droplet outlet and the substrate and heating the area between the outlet and the substrate to convert the droplets to deposited material (see figures, abstract, pages 1, 2, 4, 5, 7, 10, 17 and Table 1). Choy et al does not teach generating a flame from a burner coaxial with the droplet outlet. Blackwell et al taught combustion processes and apparatus for atomized liquid reactants wherein atomization can be done electrostatically (see col. 8, lines 27-41; col. 8, line 66 to col. 10, line 7 and figure 4). It would have been obvious to a person of ordinary skill in the art at the time the invention was made to use the fuel/oxygen flame from Blackwell et al as the heat source in Choy et al because it would have been a functional equivalent to the heat source of Choy et al in that Blackwell et al teaches that it was well known to do so for silica production. Likewise, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to apply an electric field in the arrangement of Blackwell et al because Choy et al taught that it would guide the droplets to the substrate more efficiently. Disclosure of electrostatic atomization in Blackwell et al would also have suggested the use of an electric field in the area of the burner of Blackwell et al.

11. Choy et al teaches introducing inert gas around the precursor atomizer, but does not teach the claimed sequence of annular gas jets. Blackwell et al taught the use of inert shield gas between liquid precursor droplets and an outer, annular fuel gas jet. It would have been obvious to a person of ordinary skill in the art at the time the invention was made to arrange the coaxial burner conduits and gas jets as claimed because Blackwell et al taught that it would have prevented the premature deposition of oxide material on the burner face.

Art Unit: 1731

12. Blackwell et al does not state that the flame is annular, per se. Since Blackwell et al teaches that the fuel is introduced only through the outermost concentric opening shown in figure 4, the combustion zone within the flame of Blackwell et al is necessarily annular. Blackwell et al also states that “burner buildup” is to be avoided by introducing inert gas around the feedstock. This is a clear teaching that the combustion, chemical reaction and feedstock decomposition is meant to take place away from the burner face.

13. Furthermore, a person of ordinary skill in the art would recognize that the relative gas flow rates through the channels of Blackwell et al would have determined the shape of the flame. It would have been further obvious to a person of ordinary skill in the art at the time the invention was made to adjust the relative gas flow rates through the concentric channels of Choy et al and Blackwell et al to produce an annular flame.

14. With regard to claims 33, 34, 37-44, 50 and 52-56, text portions of Choy et al are reproduced below with emphasis added and pertinent figures to show that the claimed features were disclosed by Choy et al:

Choy et al (abstract, pages 1,2,4,5, 7, 10 and 17)

Abstract :

A method of depositing a material onto a substrate comprises the steps of: (a) feeding a material solution comprising one or more precursor compounds, a solvent and a pH-modifying catalyst to an outlet to provide a stream of droplets of the material solution; (b) generating an electric field to electrostatically attract the droplets from the outlet towards the substrate; and (c) providing an increase in temperature between the outlet and the substrate.

Pages 1 and 2 :

## FILM OR COATING DEPOSITION AND POWDER FORMATION

This invention relates to film or coating deposition and powder formation.

**Ceramic**, polymer and other films coatings and powders are used in, for example, structural and functional electronic applications.

As background, the distinction between a film and a coating is very loosely defined and is not important to the operation or description of the present invention.

In one definition, a film would be considered as a layer under about 10 $\mu$ m thick, and a coating would be of greater thickness. In the present description, however, the terms are generally used interchangeably.

The following are examples of previously proposed techniques for generating such films, coatings and powders: physical vapour deposition (PVD) (a generic term given to a variety of sputtering techniques such as radio frequency (RF) sputtering, reactive magnetron sputtering and ion beam sputtering); flame spray deposition (FSD); theso-called sol-gel process; electrostatic spray pyrolysis (ESP); and chemical vapour deposition (CVD).

However, none of these techniques has been found to provide good control of the stoichiometry morphology, microstructure and electrical properties of multicomponent oxide films and a relatively high growth rate and deposited area of a deposited film. Also, the techniques tend to need expensive equipment and highly skilled technicians for effective operation.

This invention provides a method of depositing a material onto a substrate, the method **comprising the steps of:**

**(a) feeding a material solution comprising one or more precursor compounds, a solvent and a pH-modifying catalyst to an outlet to**



provide a stream of droplets of the material solution,

(b) generating an electric field to electrostatically attract the droplets from the outlet towards the substrate; and

(c) providing an increase in temperature between the outlet and the substrate.

This invention also provides apparatus for depositing films on a substrate, the apparatus comprising:

(a) an outlet for providing a stream of material solution droplets. the material solution comprising one or more precursor compounds, a solvent and a pH-modifying catalyst;

(b) means for generating an electric field to electrostatically attract the droplets from the outlet towards the substrate; and

(c) a heater for heating the substrate and providing an increase in temperature between the outlet and the substrate.

Further respective aspects of the invention (to which the various preferred features are equally applicable) are defined in the appended claims.

Embodiments of this method, which will be referred to hereinafter as electrostatic spray assisted vapour deposition (ESAVD), enable the fabrication of both thin and thick films. The technique combines the advantages of CVD and electrostatic spray deposition whilst alleviating the problems associated therewith. In comparison to other film deposition techniques, ESAVD has a high deposition rate and efficiency, and allows easy control of the stoichiometry and microstructure of the deposits. In addition, it is a simple, cheap, and low-temperature synthesis method suitable for the fabrication of a variety of different films. The method also allows the deposition of a film on large surface area substrates.

Art Unit: 1731

The use of the pH-modifying catalyst (which can be acid or alkali) can provide a clearer solution with increased electrical conductivity, and so can give finer droplets and thus a better coating quality.

The method can be performed in so that the substrate and other pieces of apparatus are open to the surrounding ambient atmosphere, the other ambient gaseous reactants refer to any other gaseous reactants (such as oxygen, for example) that may be present in the atmosphere. In another embodiment, the method may performed within the confines of a container and said other ambient gaseous reactants may be supplied to said container, thereby to enable the deposition of a particular film.

Both simple and multicomponent ceramic oxide films have been fabricated using the above mentioned method. In one embodiment, the film may be a ceramic film such as PZT (Lead Zirconate Titanate) or a doped film such as YSZ (Yttria Stabilised Zirconia). Other films may include  $\text{PbTiO}_3$ ,  $\text{BaTiO}_3$ ,  $\text{La}(\text{Sr})\text{MnO}_3$ ,  $\text{NiO}$  YSZ,  $\text{SnO}_2$ ,  $\text{In}_2\text{O}_3$  and other Indium-Tin Oxide films. The film may also be a

#### Pages 4 and 5:

Figure 5 is a flow chart illustrating steps in a polymer deposition process;

Figures 6a and 6b illustrate X-ray diffraction patterns for polymer films produced by two process variants::

Figures 7a and 7b illustrate transmittance infra-red spectra for polymer films produced by the two process variants;

Figures 8a and 8b illustrate surface reflectance infra-red spectra for polymer films produced by the two process variants;

Figures 9a and 9b are schematic diagrams showing dipole orientation in polymer films produced by the two process variants;

Figure 10 schematically illustrates a third embodiment of apparatus for material deposition;

Figure 11 illustrates a fourth embodiment, used for powder deposition;

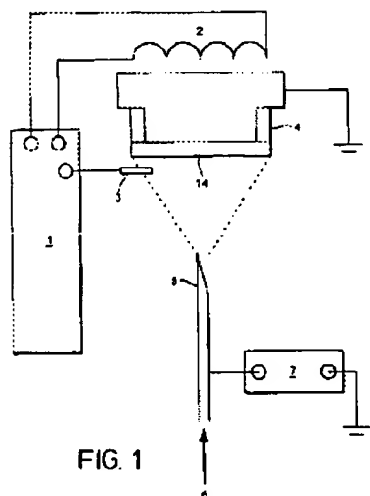
Figure 12 (curves a and b) show x-ray diffraction patterns for the nanopowders produced at 500°C and

Art Unit: 1731

800°C respectively; and

Figure 13 and 14 show the microstructures of YSZ nanopowders at different reaction temperatures.

In a first embodiment a coating solution is deposited to form a ceramic film on a substrate.



A film deposition apparatus as shown schematically in Figure 1 comprises an outlet (e.g. a nozzle, spray or similar) 5 connected to a high voltage DC source 7 preferably variable in the range 0-30 kV. In operation, a voltage of between 5 and 30 KV would be normally used for the apparatus as shown. A substrate holder 4 is earthed and is heated by a heater 2. The temperature of the substrate holder 4 is controlled by the controller 1 and an attached thermocouple 3.

Heating the substrate holder also causes a corresponding heating of the area surrounding the substrate 14 and between the substrate holder and the outlet 5. This heating serves to set up a temperature gradient whereby the ambient temperature increases as the substrate is approached from the direction of the outlet. This increasing temperature facilitates a chemical vapour reaction (see Figure 3) of the coating solution that enables deposition of the ceramic film.

When an electric field of sufficient strength is applied to the outlet 5, a corona field is produced

Art Unit: 1731

**from the tip of the outlet 5. A ceramic coating liquid is used to form the films and is fed into the outlet 5 in the direction indicated by an arrow 6.**

The outlet's inner diameter can vary from 1mm (millimeter) to 0.1mm. This relatively large inner diameter reduces the chances of clogging problems when high viscosity solutions are passed through the outlet 5.

A substantially constant flow in the range of 0.4 - 60 ml/h (millilitres per hour) is maintained by use of a syringe pump or a constant static pressure feed.

**In this way, the electrostatic field set up between the charged outlet 5 and the earthed substrate holder 4 serves to guide charged coating solution droplets to the substrate 14. Droplets of the coating solution are provided with a positive charge by way of the high voltage DC source 7. These positively charged droplets are attracted to the earthed substrate holder 4. (As an alternative, of course, the droplets could be negatively charged with an earthed holder 4, or vice versa).**

Figure 2 schematically illustrates steps in the preparation of one type of coating solution for the deposition of YSZ (Yttria Stabilised Zirconia). First, a precursor compound (in this case,  $\text{Zr}(\text{OC}_4\text{H}_9)_4$ ) is mixed with a solvent (in this case, Butanol  $\text{C}_4\text{H}_9\text{OH}$ ). This solution is stirred and a second precursor compound  $\text{Y}(\text{O}_2\text{C}_4\text{H}_9)_3$  (more generally, a metal alkoxide or an organometallic compound) is added under action of heat. The mixture is then catalysed to form a coating solution of the desired pH. In this case, ethanoic acid ( $\text{CH}_3\text{COOH}$ ) is used as a catalyst, but other acids (such as  $\text{HCl}$ ) or alkalis (such as  $\text{NH}_3$ ) may be used in the preparation of alternative coating solutions. In the case of acid catalysed reactions, the desired pH may be between 2 and 5. In the case of alkali catalysed reactions, the desired pH may be between 9 and 12.

The coating solution, a mixture of  $\text{Zr}(\text{OC}_4\text{H}_9)_4$ , Butanol and  $\text{Y}(\text{O}_2\text{C}_4\text{H}_9)_3$ , is then passed to the outlet 5

and discharged towards the substrate 14.

Preferably, the concentration of the desired coating solution is approximately 0.01-0.5 mol/l (moles per litre). In addition, the coating solution may have a viscosity in the region of about 0.01 to 50 mPa.S (millipascal seconds) depending on the type of film that it is desired to produce.

Table 1 shows the compositions of coating solutions for various deposited films.

Page 7:

In table 1, the composite precursors with alkoxide precursors are so-called **"sol" precursors**<sup>1</sup> The precursor compounds are mixed in relative quantities appropriate to the desired stoichiometry of the desired film. Sufficient catalyst is added to give the coating solution the required pH.

Ethanoic Acid is a preferred catalyst to provide a clear solution, an improved solution conductivity and therefore finer spray droplets. However, other acids and or alkalis such as hydrochloric acid, ammonia, nitric acid, Lewis acid or sodium hydroxide would all be suitable catalysts.

Acid or base catalysts can influence both the hydrolysis and condensation rates and the structures of the condensed products. Acids serve to protonate negatively charged alkoxide groups, enhancing the reaction kinetics and eliminating the requirement for proton transfer within the transition site. Therefore, acid-catalysed condensation is directed preferentially towards the ends rather than the middles of chains, resulting in more extended, less highly branched polymers. Alkaline conditions produce strong nucleophiles via deprotonation of hydroxo ligands.

Base-catalysed condensation (as well as hydrolysis) should be directed toward the middles rather than the ends of chains, leading to more compact, highly branched species. Hence, if porous films of good quality (e.g. adhesion and porosity) are required, alkalis are preferred as catalysts.

Similarly, various other inorganic or organic solvents can be used such as alcohol, water, or a mixture of alcohol and water could be used.

**Figure 3 schematically illustrates the principle of electrostatic spray assisted vapour deposition of a film from a coating solution.**

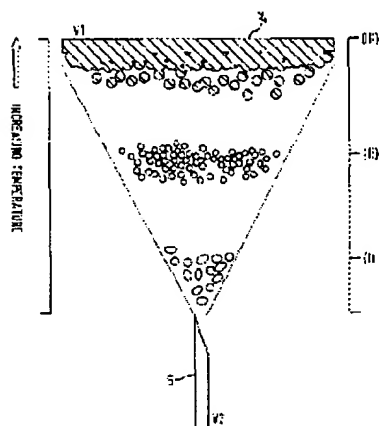


FIG. 3

The temperature preferably increases, on passing from the outlet to the substrate, from about 100°C to between 400 and 650°C. At point (I) on Figure 3, the coating solution forms a corona spray, the droplets of which are charged to a positive potential. As the droplets are attracted to the substrate 14 they begin to form clusters

<sup>1</sup>The term **Sol-gel** is used to describe chemical processes in which polymeric gels are formed from metallo-organic starting solutions (see for example: "Sol-gel Science" by C.Jeffrey Brinker and George W. Shearer, published in 1990).

Art Unit: 1731

thickness can be improved by rotating and/or translating (in general, moving) the outlet and/or the substrate holder during deposition to vary the relative positions of the outlet and the substrate with time. If rotary motion is used, this could involve, for example, rotating the substrate (which might be three-dimensional) about an axis passing through the substrate, or possibly rotating the outlet (or outlets, if more than one is used) about an axis which is not coaxial with the outlet's axis (i.e. "circling" the outlet around).

In another feature, the polarity of the electric field applied between the outlet and the substrate holder can be reversed from time to time during the deposition process. This can be beneficial to avoid the accumulation of charges (which can counteract the effect of the applied field), thus allowing thicker coatings to be produced.

"Graded" coatings can be produced by varying the concentration and/or composition of the precursor solution during deposition. (Simply, this can be achieved by depositing the contents of a first container (bottle) of precursor solution, and then switching to another container and so on).

Similarly, it has been found that films may be produced of a thickness varying from a nanometre to approximately 100 micrometers in thickness (or much thicker).

The coatings can be used in microscale circuitry or for much bulkier items such as turbine blades for jet engines, by scaling up the apparatus (the apparatus of Figures 1 and 4 as shown has a substrate diameter of about 20mm).

Single crystal substrates can be used to obtain oriented or epitaxial films. A range of microstructures including epitaxial, columnar and equiaxial growth are possible by varying the processing conditions.

The substrates may be conductive (e.g. metal) or non-conductive (e.g. glass, polymer or ceramic).

Another embodiment of an electrostatic spray assisted vapour deposition (ESAVD) apparatus is schematically illustrated in Figure 4.

The apparatus of Figure 4 is similar to that of Figure 1, except that a shaped substrate holder 104 projects towards the outlet 5 at either side of the substrate 14'

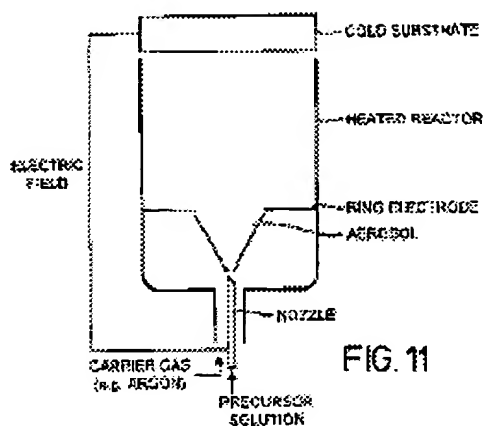
The substrate holder 104 is heated as before, and this heating serves to set up a temperature gradient whereby the ambient temperature increases as the substrate 106

Page 17:

rather than films or coatings. The example to be described is that of YSZ powders, but many other materials (particularly the materials described above with reference to the film or coating techniques) could be used.

**The differentiation between film (coating) production and powder production, using basically the same apparatus, is mainly one of temperature (although other operational parameters can be varied). If the ambient temperature between the outlet and the substrate is increased then the droplets of coating solution will tend to form powder particles before hitting the substrate. This effect can be exaggerated by slowing down the flight of the droplets - e.g. by changing the flow rate or the electric field - to give more time for the powder to form. Alternatively, if a cold substrate is used, then the droplets will condense into powder particles on hitting the substrate.**





**Figure 11 illustrates a suitable apparatus for manufacturing the powdered YSZ.** The apparatus comprises a tubular, up-flow reactor equipped with an external resistive heater. YSZ sol precursor is delivered at an appropriate flow into a stainless steel capillary outlet (100 $\mu$ m inside diameter, 650 $\mu$ m outside diameter) which is maintained at a dc voltage of 10-30kV (positive polarity). The capillary electrode is placed 15-30 mm from a ring electrode maintained at 1kV dc voltage. The function of the ring electrode is to focus the spray aerosol into the reactor chamber and select the appropriate nanosized particles to reach the substrate. The high local electrostatic field at the capillary tip causes an emission of charged aerosol from the highly deformed liquid interface. **A stream of Argon gas from a coaxial glass capillary (800 $\mu$ m inside diameter) carries the spray charged aerosol through the ring electrode into the reaction zone of the reactor, finally to arrive at the cold substrate (having a negative polarity).**

The YSZ sol precursor with monodispersed nanosize particles is synthesized as follows.

First,  $Y(NO_3)_3 \cdot 6H_2O$  is dissolved in 2-propanol while stirring for about 15 min at room temperature yielding a clear solution. Zirconium tetra n-butoxide,  $Zr(OC_3H_9)_4$  is added into the previous solution according to the stoichiometry of a desired final composition of  $(ZrO_2)_{0.92}(Y_2O_3)_{0.08}$ . The final sol precursor concentration and pH are preferably 0.05M and 3-5 respectively. The hydrolysis and condensation can be carried out at room temperature under stirring.

The YSZ nanoparticles produced by a prototype of this technique were

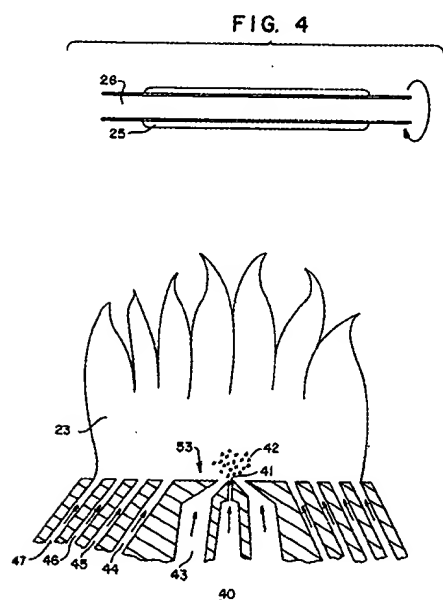
15. With regard to claims 30-32, 35, 36 and 46-49, portions of Blackwell et al with emphasis added and pertinent figures are reproduced below:

Blackwell et al Col. 8, lines 27-41:

Several types of atomizing means capable of forming very small particles of liquid are known in the atomization art as disclosed in Atomization and Sprays, by Arthur H. Lefebure, Hemisphere Publishing co., 1989, which is incorporated herein by reference. Atomizers can be operated by various energy sources such as liquid, gas, mechanical, electrical and vibrational, and may be categorized as, for example, jet, swirl, jet-swirl, pneumatic, rotary, acoustic, ultrasonic, and **electrostatic**. Preferably, a jet atomizer is used; even more preferably, the jet atomizer is a swirl-jet atomizer, which swirls the liquid and then, as atomizers generally do, squirts the liquid at high velocity out of a small orifice. Various types of atomizers are discussed in Liquid Atomization, by L. Bayvel and Z. Orzechowski, Taylor & Francis, (1993), which is hereby incorporated by reference.

Blackwell et al Col. 8, line 66 to col. 10, line 7:

FIG. 4 is a cross-sectional view of a preferred embodiment of the apparatus of the present invention. Here, burner 40 incorporates within its structure atomizer 41, which injects very finely atomized liquid reactant particles into flame 23. As with the previously described embodiments, amorphous soot 25 produced by combustion of the liquid reactant is collected on rotatable mandrel 26.



As shown in FIG. 4, burner 40 comprises a series of concentric channels surrounding atomizer 41. Liquid siloxane is delivered through atomizer 41. A stream of an inert gas such as nitrogen gas, a mixture of oxygen gas and nitrogen gas, or oxygen gas alone delivered through channel 43 atomizes the liquid feedstock by the kinetic energy of the flowing gas to create liquid projections 42 which are converted into soot reactant particles in burner flame 23. The area proximate to the burner face 53 and flame 23 thus acts as a conversion site for converting liquid projections 42 into soot reactant

Art Unit: 1731

**particles.** oxygen gas may be delivered to flame 23 through channels 45 and 46.

**An inert gas, such as nitrogen, argon or helium is delivered through channel 44 to inhibit reaction of the liquid feedstock and soot build-up on burner face**

53. Applicants have found that when oxygen or a mixture of oxygen and inert gas is used as the atomizing gas, better results are obtained by delivering an inert gas through channel 44. **A premix of oxygen and a fuel such as methane is conducted to the flame through outermost channel 47.** A burner fitted with an atomizing injector, such as the embodiment depicted in FIG. 4, produces a wide soot stream, which achieves improved concentricity of the core and cladding regions of a subsequently formed optical waveguide fiber.

The most preferred burner 40 of the invention as shown in FIG. 4 is comprised of a pneumatic atomizer. With such a pneumatic atomizer, the liquid siloxane feedstock delivered through atomizer 41 is atomized by the kinetic energy of a flowing gas stream through inner most channel 43. High velocity gas is utilized in atomizing the feedstock. This produces atomized liquid projections 42 with a velocity in the range of 0.5 to 50.0 m/sec. The use of an inert gas such as N.sub.2 gas is preferred with the pneumatic atomizer. Use of N.sub.2 gas as the pneumatic gas helps to blanket the feedstock from the oxygen in the flame and prevents burner buildup.

To reduce the velocity of the atomizing gas and prevent surface defects on the soot blank, O.sub.2 gas is the most preferred gas to be used with the pneumatic atomizer. It was previously believed that O.sub.2 would not help to prevent combustion of the feedstock prior to full vaporization of the liquid feedstock. However, applicants have discovered that using oxygen as the atomizing gas allows better mixing of the siloxane with oxygen before

Art Unit: 1731

conversion to soot. It is believed that use of this atomizing gas results in quicker heating of the liquid and helps provide the  $O_{2}$  needed for the reaction. Therefore, the velocity of the oxygen atomizing gas can be significantly lowered, at least by about 50% of the velocity of nitrogen atomizing gas. This reduction in gas velocity consequently reduces burner flame turbulence and soot blank defects.

It was also observed that using a mixture of oxygen and nitrogen as the atomizing gas allowed for lower atomizing gas velocity and reduced blank defects. Preferably, the atomizing gas employs greater than 50% oxygen. For example, a mixture of 75% oxygen gas and 25% nitrogen gas by volume as the atomizing gas enabled a significant reduction in atomizing gas velocity and a reduction in soot blank defects. However, compared to this example, using oxygen alone provided for a lower atomizing gas velocity.

With the use of the pneumatic atomizer in the invention, the high velocity blast gas is deployed in an effective manner to achieve a beneficial level of atomization of the siloxane at the burner and in the flame.

In practicing the invention, even though it is preferred to have atomizer unit 41 as an integral part of the burner unit 40, it is possible to use a pneumatic atomizer which is spatially separated from the burner, as with atomizers 21 and 31 in FIG. 2 and FIG. 3.

16. In claim 30, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to expect the chemical reaction and/or decomposition of the precursors

Art Unit: 1731

to occur in the overlap zone between the droplet stream and the annular flame because the droplets would be forced to travel through the inert gas stream and to reach the flame of Blackwell et al.

17. In claim 31, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to expect the fuel flow to be diverging because a gas ejected from an orifice under pressure diverges.

18. In claim 32, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to make the first (droplet) and second (fuel) outlets coaxial as in Blackwell et al because Blackwell et al taught that this permitted the use of an integral atomizer.

19. In claim 35, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to introduce a “cold gas” from a third outlet between the droplets and the fuel because Blackwell et al taught that this prevented premature deposition of oxide material on the burner face.

20. In claim 36, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to make the first (droplet) and third (cold gas) outlets coaxial because Blackwell et al taught that this made the feature of claim 35 possible.

21. In claim 46, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to expect the chemical reaction and/or decomposition of the precursors to occur in the overlap zone between the droplet stream and the annular flame because the droplets would be forced to travel through the inert gas stream and to reach the flame of Blackwell et al.

Art Unit: 1731

22. In claim 47, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to make the first (droplet) and second (fuel) outlets coaxial as in Blackwell et al because Blackwell et al taught that this permitted the use of an integral atomizer.

23. In claim 48, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to introduce a “cold gas” from a third outlet between the droplets and the fuel because Blackwell et al taught that this prevented premature deposition of oxide material on the burner face.

24. In claim 49, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to make the first (droplet) and third (cold gas) outlets coaxial because Blackwell et al taught that this made the feature of claim 48 possible.

25. Claims 29-44 are rejected under 35 U.S.C. 103(a) as being unpatentable over Choy et al in view of Hitachi (JP 56-5337).

26. Choy et al taught methods for depositing ceramic material as outlined in the above rejections. Choy et al did not teach generating an annular flame.

27. As clearly illustrated in Figure 1 of Hitachi, the flame 16' is annular. Hitachi did not teach that a 'fuel' was delivered from a second outlet about the atomized raw material stream. Since the raw material was atomized with hydrogen and oxygen was introduced from a coaxial outlet surrounding the hydrogen and raw material stream, the oxidant can be considered fuel. Regardless, the figure clearly shows an annular flame 16'. It would have been obvious for a person of ordinary skill in the art to use the flame from Choy et al as the heat source in Choy et al because it would have been a functional equivalent to the heat source of Choy et al in that Hitachi taught that it was well known to do so for silica production.

Art Unit: 1731

28. Choy et al taught introducing inert gas around the precursor atomizer, but did not teach the claimed sequence of annual jets. Hitachi taught the introduction of Ar, He, N<sub>2</sub> or Ne (ref. no. 8) through conduit 7 (see page 238, (3): lines 11 and 12). It would have been obvious for a person of ordinary skill in the art to deliver a flow of 'cold gas' from a third outlet around the raw material stream and within the flow of fuel (oxidant) because Hitachi clearly illustrated that such an arrangement would help produce an annular flame 16'.

29. In claim 30, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to expect the chemical reaction and/or decomposition of the precursors to occur in the overlap zone between the droplet stream and the annular flame because the droplets would be forced to travel through the inert gas stream and to reach the flame of Hitachi.

30. In claim 31, It would have been obvious to a person of ordinary skill in the art at the time the invention was made to expect the fuel flow to be diverging because a gas ejected from an orifice under pressure diverges. Furthermore, Hitachi's flame shape is clearly suggestive of a diverging flow.

31. Claim 51 is rejected under 35 U.S.C. 103(a) as being unpatentable over Choy et al and Blackwell et al as applied to claim 45 above, and further in view of Japan 62-220376 (see abstract and figures).

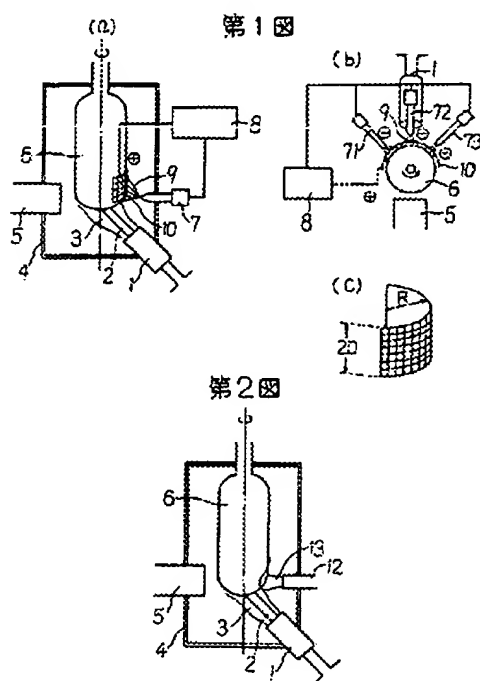
32. Choy et al and Blackwell et al do not teach a mesh disposed between the first outlet and the substrate. Japan '376 taught:

PURPOSE: To efficiently obtain the title preform without cracking by cooling a porous preform with an ion jet generated by a corona discharge, and simultaneously depositing fine glass particles on the preform at the time of producing the porous optical fiber preform by the axis formation in a vapor phase.



Art Unit: 1731

**CONSTITUTION:** A corona-discharge electrode 7 is arranged above a fine glass particle synthesizing burner 1. A glass material and a combustion gas are supplied to the burner 1 to cause a flame hydrolysis reaction, the formed fine glass particles 3 are deposited on the surface of a starting material, and a porous optical fiber preform 6 is obtained. A negative voltage is simultaneously impressed on the corona-discharge electrode 7 and a positive voltage on a wire-mesh electrode 10 from a DC power source 8 to generate a corona discharge between both electrodes, and an ion jet 9 is produced. The porous preform 6 is cooled by the ion jet 9, the fine glass particles are simultaneously deposited, and the preform 6 is produced.



33. It would have been obvious to a person of ordinary skill in the art at the time the invention was made to use a wire-mesh electrode like that of Japan '376 in the arrangement of Choy et al and Blackwell et al because a mesh would have been equivalent to the ring electrode of Choy et al.

***Response to Arguments***

34. Applicant's arguments filed December 13, 2004 have been fully considered but they are not persuasive.

35. The applicant has alleged that the method of Blackwell et al failed to produce an annular flame combustion region and the apparatus of Blackwell et al would never produce an annular flame combustion region. According to the applicant, figure 4 of Blackwell et al is sufficient to teach that the flame of Blackwell et al was a continuous flame and that the outermost channel of the Blackwell et al burner was frusto-conical. Neither term "continuous flame" nor "frusto-conical" appears in the disclosure of Blackwell et al.

36. The examiner's position with regard to the annular flame was presented in the previous office action. In the present office action, new rejections based on Hitachi are presented to bring to light the annular flame illustrated as 16' in the Hitachi figures.

37. The features of dependent claims 30-44, 46-50 and 52-56 were clearly taught or suggested by the disclosure of Choy et al and Blackwell et al, as shown in the highlighted text presented herein. Until the submission of the appeal brief, the applicant did not allege that these features were not taught. The examiner has herein demonstrated that they were in fact taught.

38. Claim 51 recited a mesh which would have been a functional equivalent to the ring of Choy et al and therefore obvious. In response to applicant's argument that the claimed mesh is not related to the presence of the electric field, the fact that applicant has recognized another advantage which would flow naturally from following the suggestion of the prior art cannot be the basis for patentability when the differences would otherwise be obvious. See *Ex parte Obiaya*, 227 USPQ 58, 60 (Bd. Pat. App. & Inter. 1985).

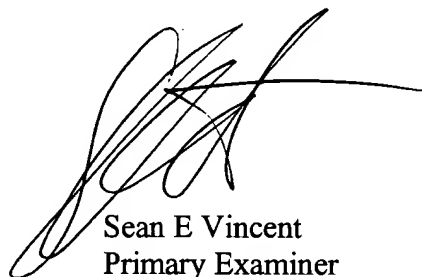
***Conclusion***

39. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Sean E. Vincent whose telephone number is (571) 272-1194.

The examiner can normally be reached on M - F (8:30 - 6:00).

40. If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Steven P. Griffin can be reached on (571) 272-1189. The fax phone number for the organization where this application or proceeding is assigned is 703-872-9306.

41. Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).



Sean E Vincent  
Primary Examiner  
Art Unit 1731

S Vincent  
April 12, 2005